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Title: Semiconductor Nanocrystals: Tiny Particles with “Quantum Powers”

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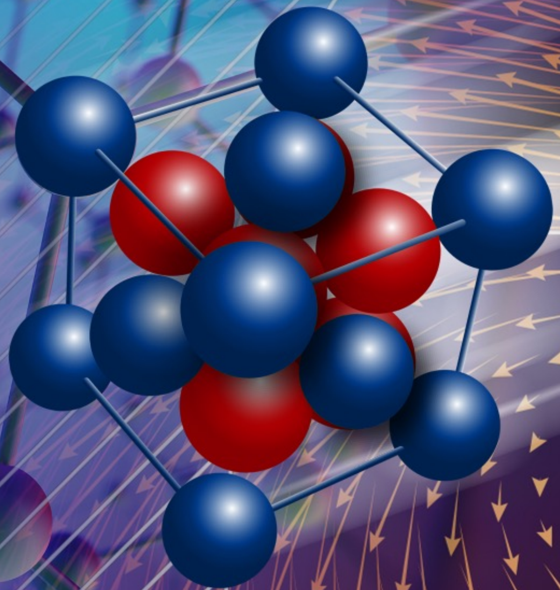
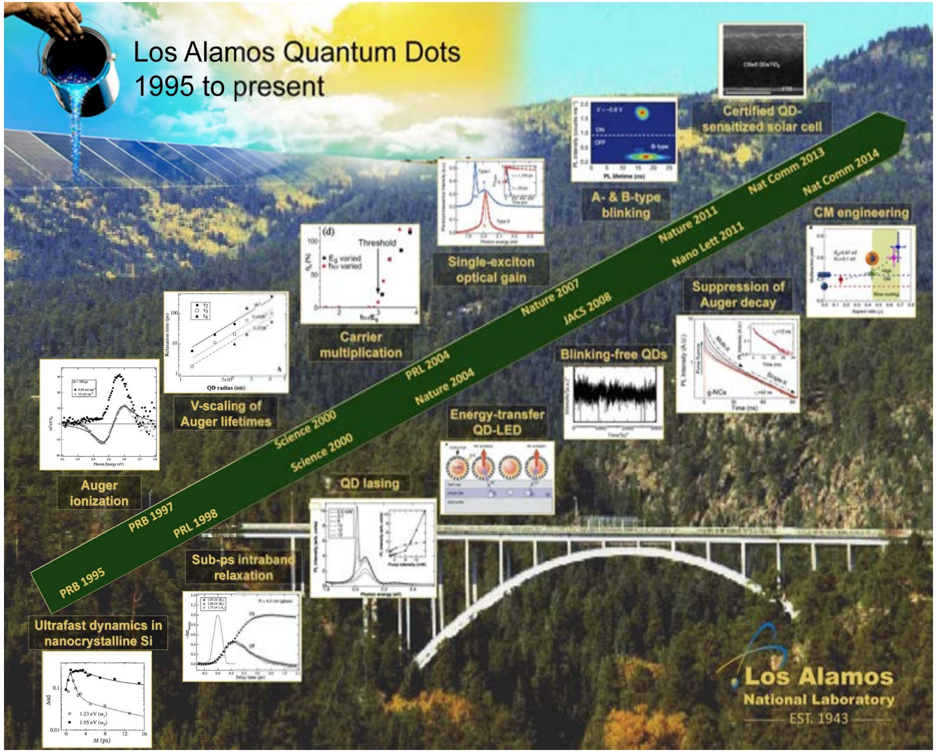
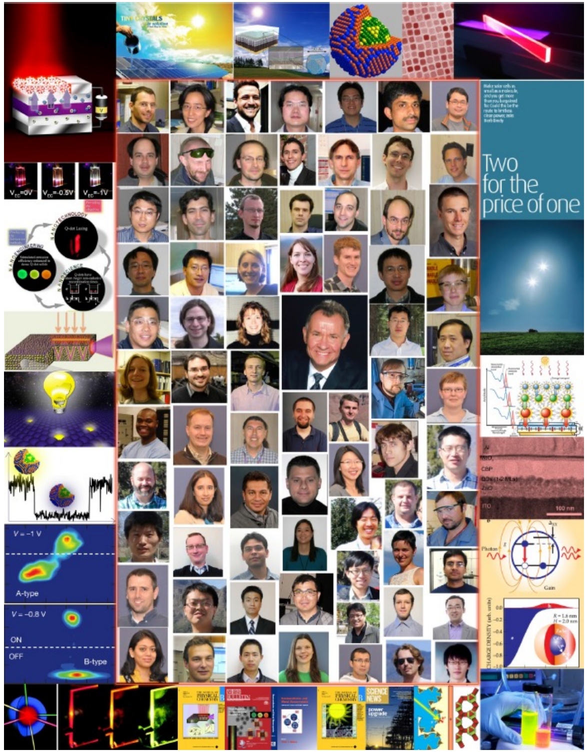
Semiconductor Nanocrystals: Tiny Particles with "Quantum Powers"

Victor I. Klimov,
Chemistry Division, C-PCS



Los Alamos Quantum Dots: Twenty Five Years of Innovation…and Fun





Materials for the Future

- Center for Integrated Nanotechnologies (CINT), DOE's NSRC (2000 - …)

Sandia National Laboratories

Los Alamos NATIONAL LABORATORY EST. 1943

- Center for Advanced Solar Photophysics (CASP), DOE's EFRC (2009 - 2019)

Los Alamos NATIONAL LABORATORY EST. 1943

NREL NATIONAL RENEWABLE ENERGY LABORATORY

THE UNIVERSITY OF CHICAGO

GEORGE MASON UNIVERSITY

Penn UNIVERSITY of PENNSYLVANIA

Materials for the Future

- **LANL's Pillar: Materials for the future**

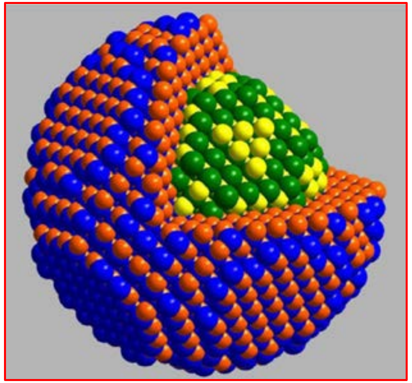
Controlled functionality through:

Quantum confinement

Wavefunction engineering

Engineered surfaces & interfaces

Controlled inter-dot coupling



- **LANL's Areas of Leadership**

Integrated nanomaterials:

Emergent functionality at the nanoscale

Next generation materials for manipulating photons

Advanced materials for optoelectronics

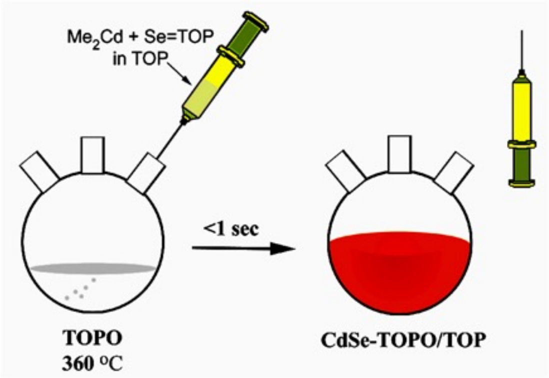
- **Overarching Science Questions:**

How to predict & control design of nanostructure and/or interfaces to achieve emergent functionalities?

How do we understand & exploit competing interactions associated with reduced dimensionality & interfaces to yield tailored properties?

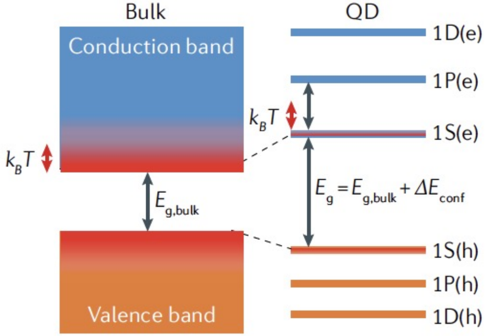
Born in Colloidal Solution: Artificial Atoms with Tunable Energies

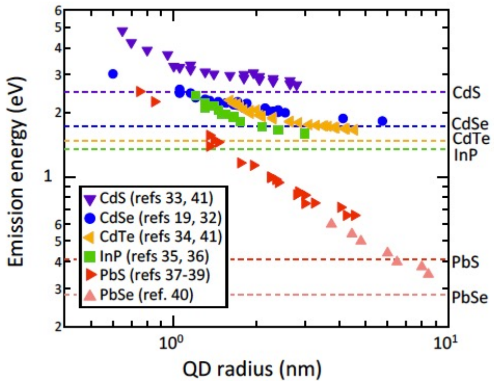
- **Fabrication: Moderate-temperature colloidal chemistry**

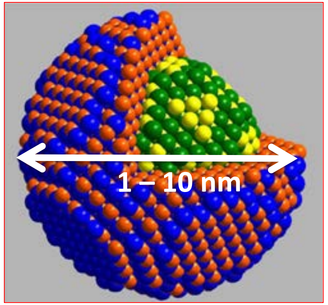


R = 10—50 Å, ΔR/R = 4—7%

- **Size-tunable electronic energies**







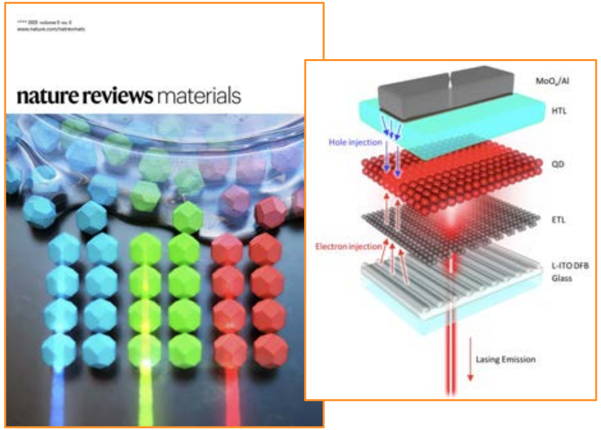
$$E_g(NQD) = E_g(bulk) + \frac{\hbar^2\pi^2}{2m_{eh}R^2}$$

quantum confinement term (> 200 meV)

Tiny Particles…with Magic Powers

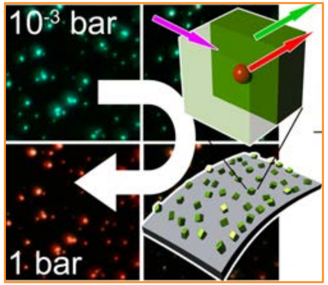
- **Lighting & Displays**

- **Lasing**

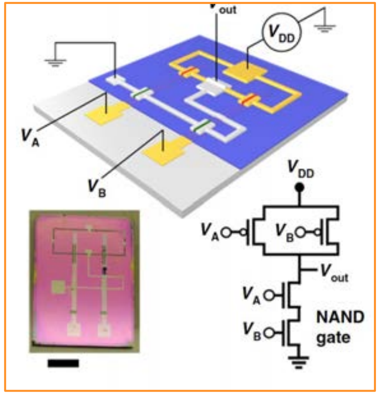


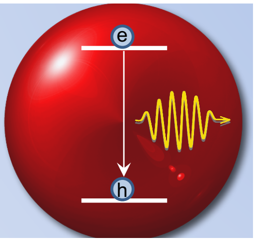
- **Solar energy**

- **Sensing**



- **Electronics**



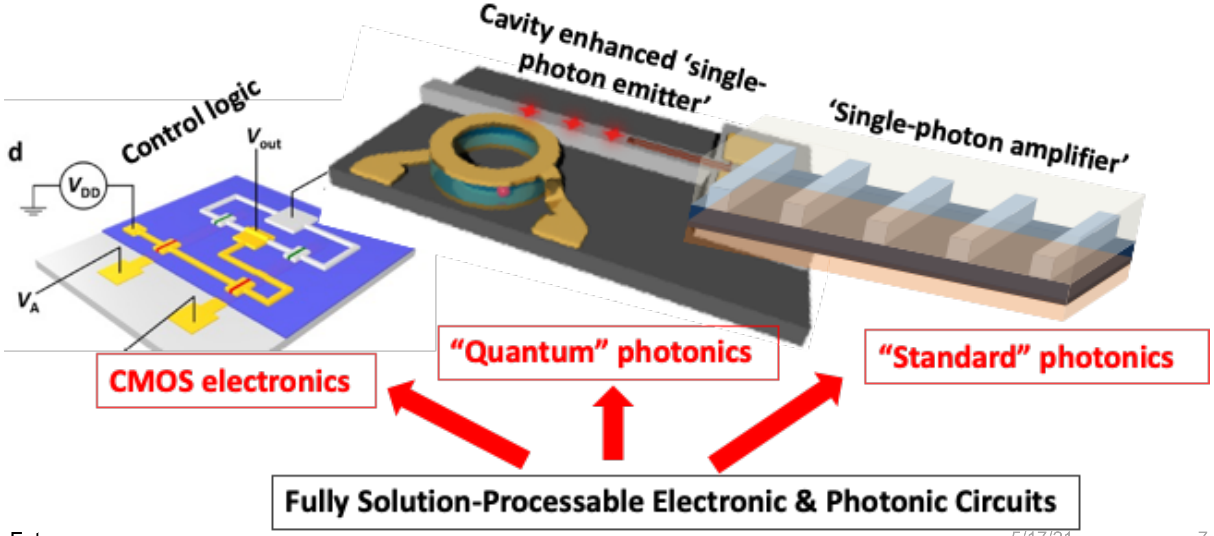


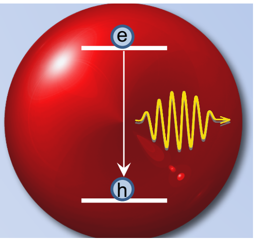
LDRD DR Project: Quantum Photonics with Semiconductor Nanocrystals

- **Overarching Objective**: To explore the utility of colloidal quantum dots as highly-versatile, solution-processible materials for implementing wavelength-selectable single-photon emitters (SPEs) complemented by on-chip light-amplification circuits, laser sources, and logic-gate devices.

- **'Single-photon emitter' (SPE):** Emits exactly **one photon at a designated time** and all photons are quantum-mechanically identical or '**indistinguishable**'

Chemistry:
C-PCS
Materials Physics & Applications:
Nat High-Magnetic Field Lab
Center for Integrated Nanotech
Nuclear Eng & Nonproliferation:
NEN-1
Theory:
T-1





Project Milestones & Timeline

Project Timeline

"Quantum light 101"

Y1 *Achieving high single-photon purity, near-infrared (NIR) wavelength, high spectral stability*

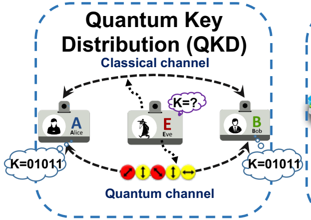
"Quantum light 201"

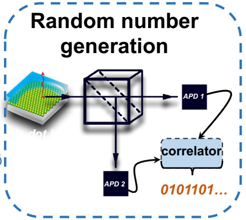
Y2 *Expanding/enhancing QL101 capabilities: electrical pumping, coupling to cavities…*

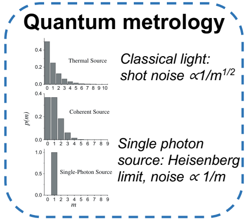
"Quantum light 301"

Y3 *Achieving high excitonic coherence, photon indistinguishability, maintained at elevated temperature*

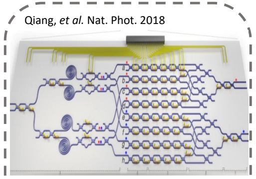
Prospective applications:
High single-photon purity



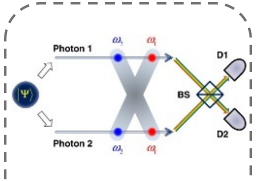




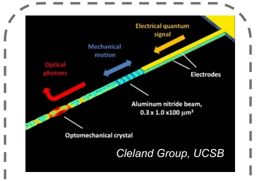
Prospective applications:
High excitonic coherence



Linear computing



Entangled photon pairs

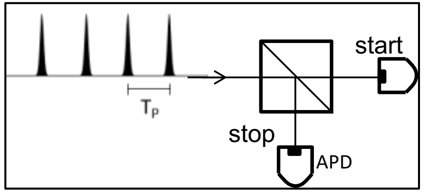


Quantum transduction

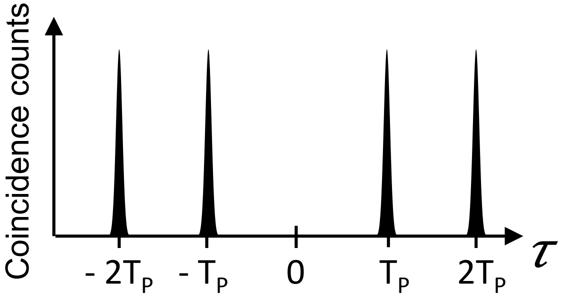
Photon Statistics & Single-Photon Purity

- **Photon statistics: Chaotic (classical), coherent (laser) & anti-bunched (quantum) light**

Hanbury, Twiss & Brown experiment



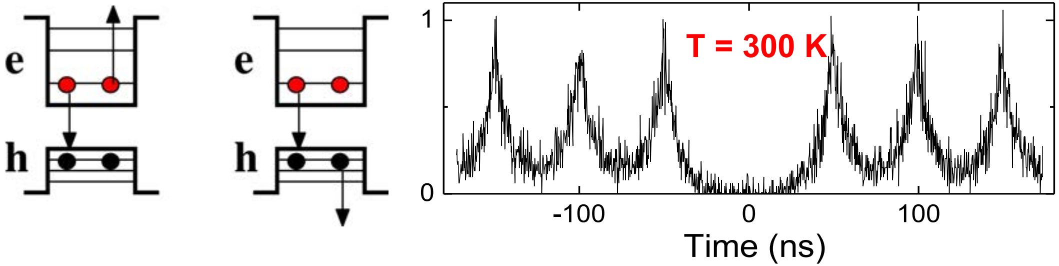
Ideal single-photon emitter: $g^{(2)}(0) = 0$

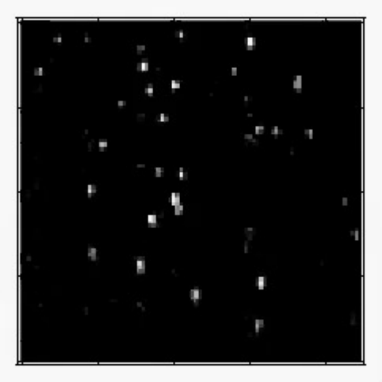


- **Antibunching mechanisms in colloidal QDs: Quantized Auger recombination**

$F_{spp} = 1 - g^{(2)}(0)/g^{(2)}(T) = 1 - Q_{XX}/Q_X = 1 - 4\tau_{XX}/\tau_X = 0.99$

V. Klimov *et al.*, *Science* **287**, 1011 (2000)

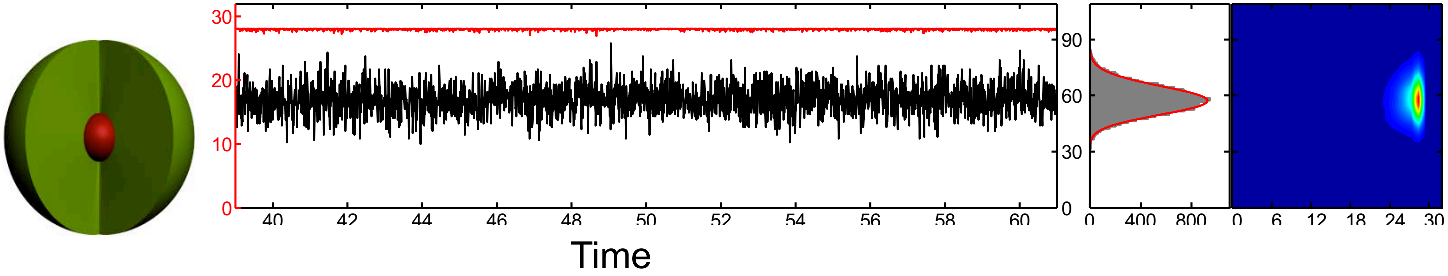




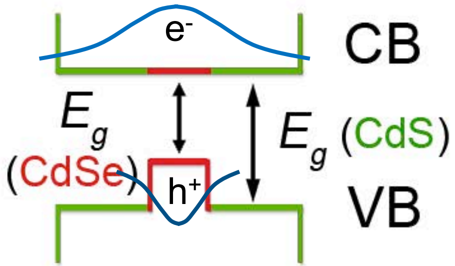
Pros & Cons of “Giant” CdSe/CdS QDs

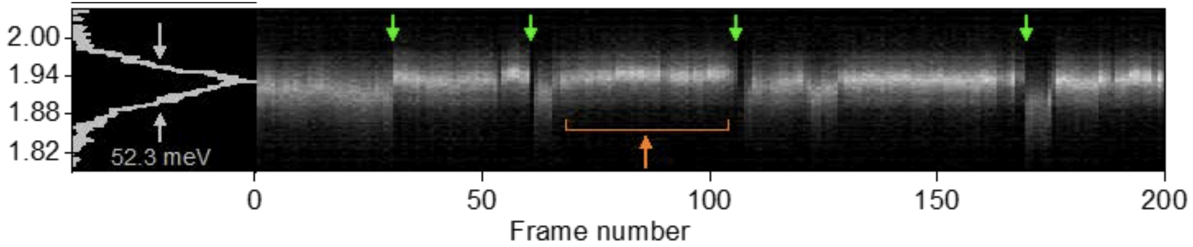
- “Giant” thick-shell CdSe/CdS QDs: Nearly complete suppression of intensity fluctuations

C. Galand, et al. *Nature* **479**, 203 (2011)



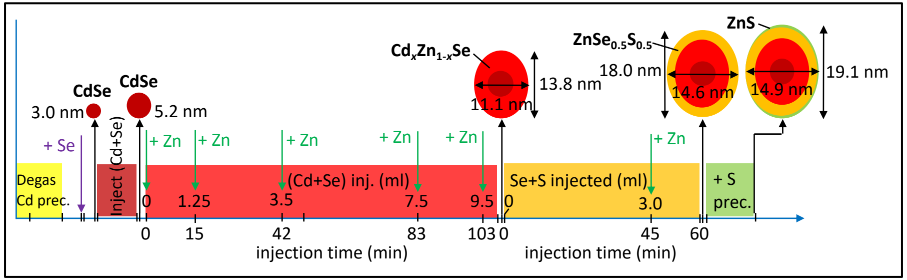
- … Still significant spectral fluctuations



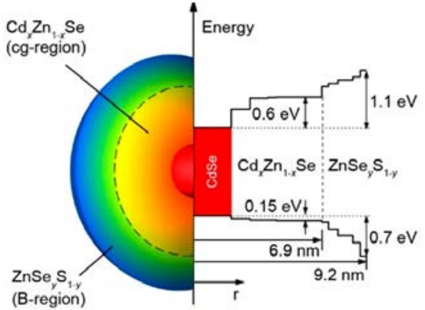


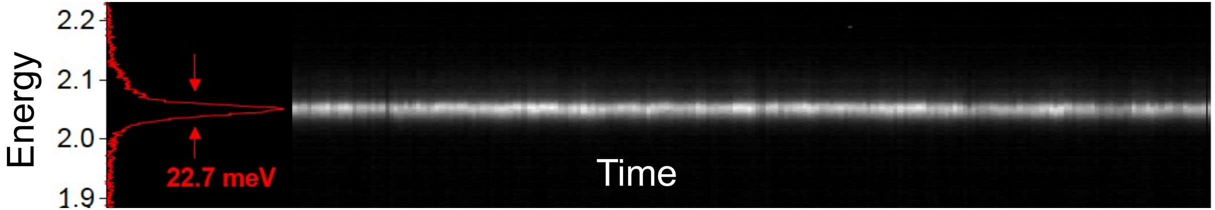
Fixing the Problem of Standard "Giant" QDs

- **Asymmetrically strained, continuously graded CdSe/ZnCdSe cg-QDs**

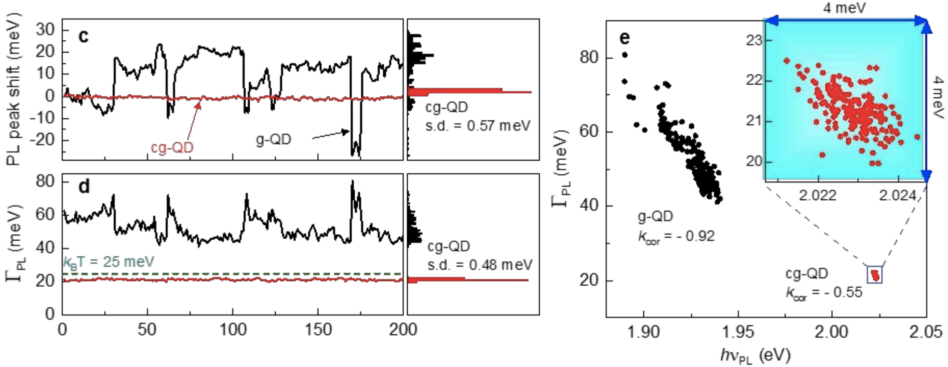


O. Kozlov… V.I. Klimov, *Science* **365**, 672 (2019)





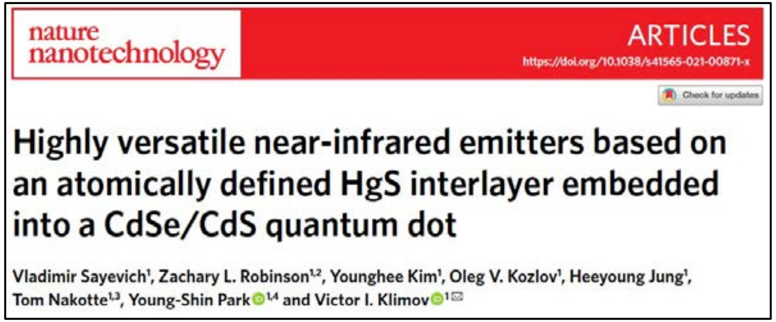
- **Sub-kT spectral fluctuations**



J. Lim, Y-H. Park, V.I. Klimov, *Nature Mater.* **18**, 249 (2019)

CdSe/HgS/CdS QDs: Near-IR Upgrade for CdSe/CdS QDs

- “Nonblinking” <u>NIR CdSe/HgS/CdS</u> QDs with atomically defined HgS interlayer


nature nanotechnology

ARTICLES

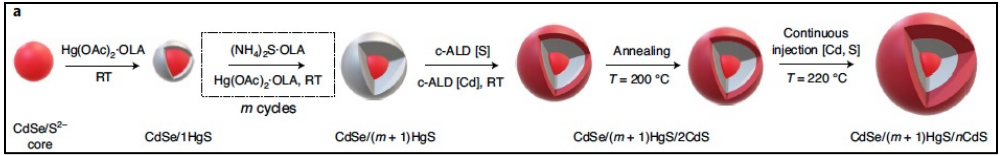
https://doi.org/10.1038/s41565-021-00871-x

Check for updates

Highly versatile near-infrared emitters based on an atomically defined HgS interlayer embedded into a CdSe/CdS quantum dot

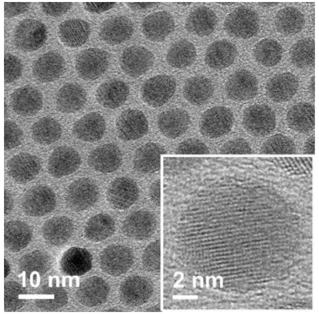
Vladimir Sayevich[1], Zachary L. Robinson[1,2], Younghee Kim[1], Oleg V. Kozlov[1], Heeyoung Jung[1], Tom Nakotte[1,3], Young-Shin Park[1,4] and Victor I. Klimov[1]

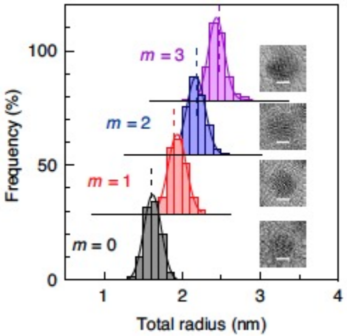
- Thermodynamically controlled L-b-L growth

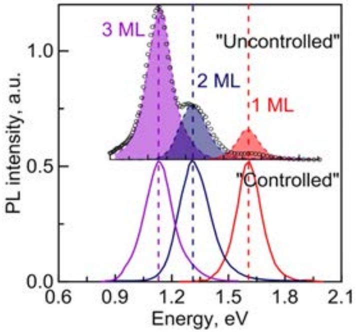


- Atomic-level control of the HgS interlayer thickness

V. Sayevich, … V.I. Klimov,
Nature Nanotech. March 25 (2021)

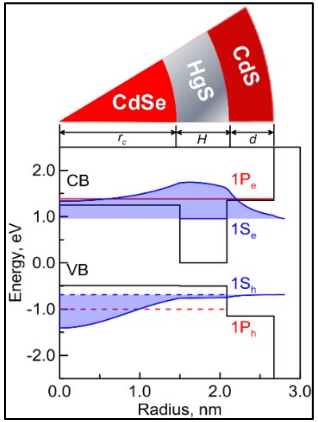


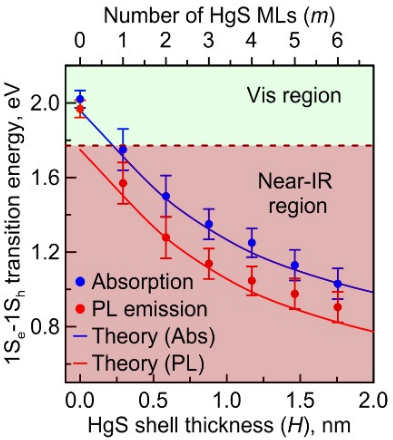




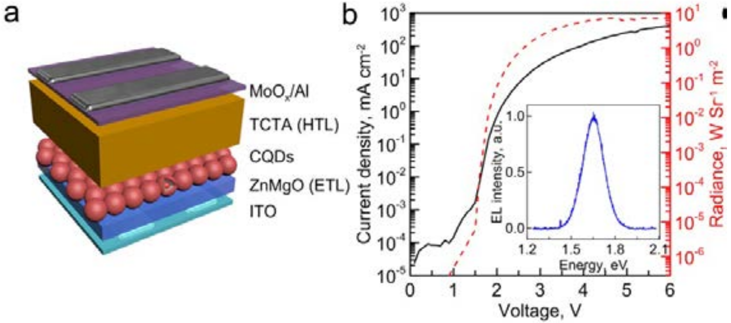
CdSe/HgS/CdS QDs: Perfect NIR Emitters

- Highly efficient, spectrally tunable NIR emitters (QY > 60%)





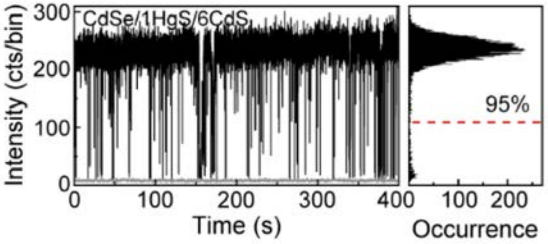
- Excellent electroluminescent materials (NIR-LEDs)



- Highly stable single-photon emitters

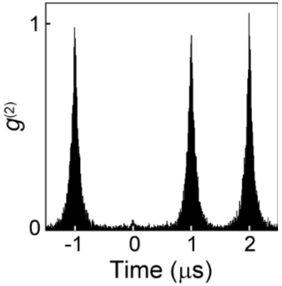
Nearly blinking-free emission

$f_{ON} > 95\%$



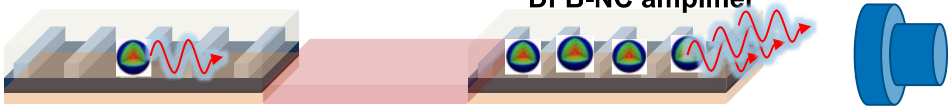
Perfect single-photon purity

$F_{spp} > 97\%$



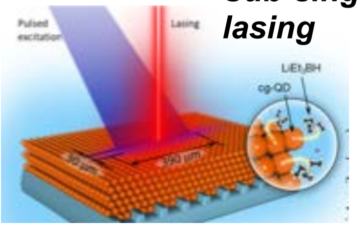
Photon Amplification

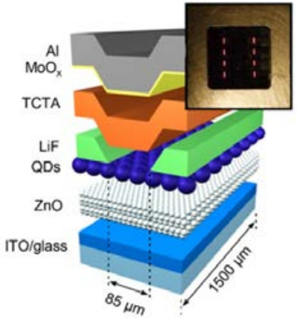
High-sensitivity detection using single-photon QD amplifiers (sub-threshold laser)



cg-QDs are excellent optical gain materials (suppressed Auger decay, large gain)

Sub-single-X lasing



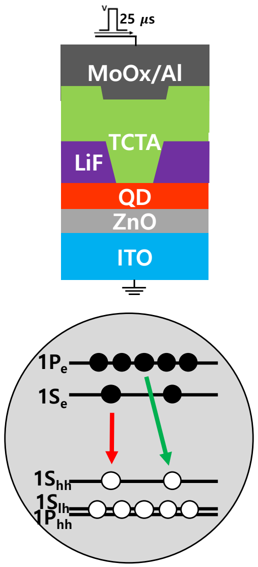


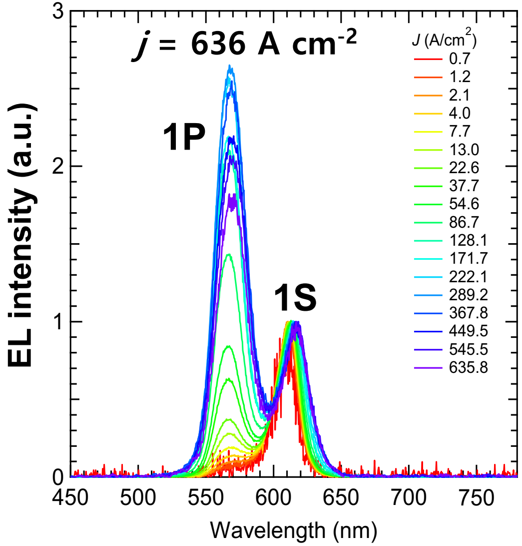
Optical gain with electrical pumping

O. Kozlov, …V.I. Klimov, *Science* **365**, 672 (2019)

J. Lim, Y.S. Park, V.I. Klimov, *Nature Mater.* **17**, 42 (2018)

Complete 1S and 1P population inversion using pulsed, current focusing QD-LEDs





H. Jung, Y.S. Park, V.I. Klimov, *Nature Phot.* **in press** (2021)

Ongoing & Future Research

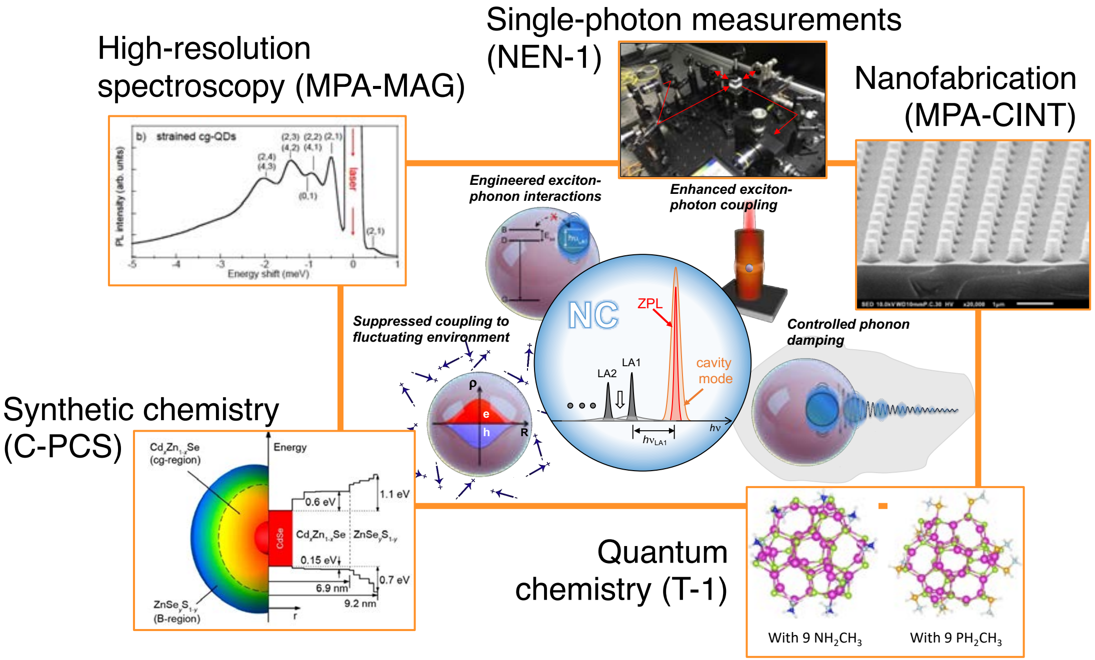
<u>Primary Challenge:</u> Achieve photon indistinguishability via control of extrinsic and intrinsic mechanisms for dephasing

Path forward

- Manipulation of NC electronic wavefunctions, surfaces, and matrices to eliminate spectral instabilities due to fluctuating environment

- Manipulation of NC electronic states, phonon spectra and matrices to suppress dephasing due to electron-phonon scattering

- Exploiting resonant coupling to optical cavities to enhance and sharpen the coherent exciton line and suppress incoherent phonon replicas.



Acknowledgements

LABORATORY DIRECTED RESEARCH & DEVELOPMENT

- **Nanotechnology & Advanced Spectroscopy Team**

- **Quantum Photonics LDRD-DR Project**

Victor Klimov (C-PCS)

Sergei Tretiak (T-1)

Sergei Ivanov (MPA-CINT)

C-PCS
MPA-MAG
MPA-CINT
NEN-1
T-1

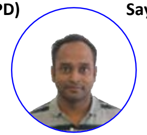
Mark Croce (NEN-1)

Scott Crooker (MPA-MAG)

Robinson (PD)

Sayevich (PD)

Kim (PD)



Ghosh (PD)

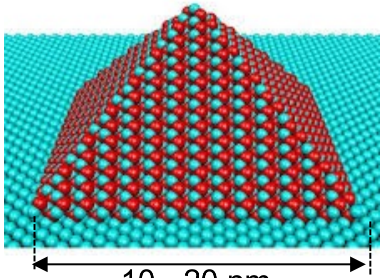
Ahn (PD)

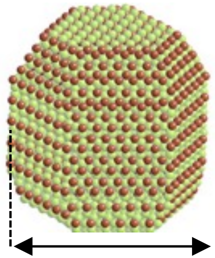
Colloidal Quantum Dots as Single-Photon Emitters

- InAs/GaAs QDs (T = 4.5 K)

  Single-photon purity = 99.1%
  Indistinguishability = 97.5%

  Dusanowski, et al., *Phys. Rev. Lett.* **122**, 173602 (2019)





- Free-standing particles: uniquely suited for applications in imaging and sensing (by integrating with, e.g., scan probes)
- Easily scalable structures prepared using non-clean room techniques (e.g., inkjet printing)
- Very inexpensive

Epitaxial quantum dots

- Small inter-level spacing: Need cryogenic-cooling (liquid helium)
- Limited range of accessible wavelengths (~800 – 1100 nm; defined by InGaAs band-gap)
- Difficulty for on-chip integration with Si circuits (lattice mismatch)
- Dot-to-dot variability in emission energy (size polydispersity)

Colloidal quantum dots

- Can serve as SPEs at elevated T: large inter-level spacing ($\gg k_B T$), strongly confined acoustic phonons
- Readily tunable emission *via* size-controlled band-gap (UV – visible – IR)
- Chemically processable particles: easy to integrate with electronic and photonic circuits
- In principle, can be prepared as monodispersed structures with identical emission energies